Please amend Figures 1, 2, 3 and 5 to add reference numeral 14 to figures 1 and 2; numeral 102 to Figure 3; and correct the printing error in Figure 5 as shown within the circles in the attached prints.

New figures incorporating these corrections are also attached for replacement of the corresponding Figures currently on file.

**Remarks**

Claims 1 to 28 inclusive are being prosecuted

By this amendment the drawings and disclosure have been revised, it is believed, to overcome the objections raised in paragraphs 1 and 2 of the Office Action and the claim language has been corrected, it is believed, to overcome all the objections raised in paragraphs 3 to 5 inclusive and to more distinctly and explicitly define the invention. The claims particularly the broadest claims (Claims 1 and 23) have been amended to more precisely define the present invention and to clearly distinguish the invention from any prior art of which Applicant is aware.

New claims 26, 27 and 28 have been added to round out the protection. **Please charge the additional fee require under Fee Code 2202 @ 9.00/claim = $27.00 to my deposit account no. 18 2150 with the US Patent Office.**

The drawings (Figures 1, 2, 3 and 5) have been amended to correct the defects noted in the Office Action and to put the proper symbols in Figure 5 which symbols apparently printed as boxes as also occurred and has already been corrected in the text.

The claims have been amended, it is believed, in a manner to overcome all the objections raised in Paragraph 5 of the office Action with the exception of the use of the term "discrete".. This term is believed to be correct based on its dictionary meaning "Disconnected from others, distinct or separate" which is the meaning that is intended to be conveyed.

The broadening of the range of the characteristic dimension to between 5 and 1000 is justified as the broader range is clearly supported by the application as filed (see for example page 10 line 5 and claim 9)

The broadest claims are is now limited to the

> "discrete Ni crystallites" *being* "formed on a support element by a several incipient wetness steps process incorporating a plurality of Ni impregnation cycles, said catalyst"

van Looij et al uses only a single Ni impregnation step and as a result the structure of the van Looij et al. catalyst is significantly different from the structure of the catalyst of the present invention.

The broadest claims are further limited to

"dispersion on said support element of no more than 0.2 square meter of exposed nickel/ square meter of support surface."

This amount of dispersion of Ni ("of no more than 0.2 square meter of exposed nickel/ square meter of support surface") as defined in the claims and as required when practicing the present invention provides an area of reducible nickel per area of catalyst $m^2/m^2$ on a % basis of 14% at optimum conditions whereas the Ni α alumina based catalyst of van Looij et al. is about 86% providing another very significant difference between the present invention and the prior art. (see the attached calculations).

It is submitted that the claims as amended overcome all the formal objections raised in the office Action and clearly distinguish the present invention from the prior art and that this application is in condition for Allowance and such action is respectfully requested.

Respectfully submitted,

C.A.Rowley Reg. No. 20,781
Agent for Applicant

Date: January 15, 2003
Telephone (613) 398 1409

**Certificate of Transmission**

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## VERSION WITH MARKINGS TO SHOW CHANGES MADE

**In the Specification**

The paragraph beginning on page 5 line 15 has been revised as follows

Broadly the present invention relates to a regeneratable Nickel (Ni) catalyst particularly suited for a hydrocarbon reforming process, said catalyst comprising discrete Ni crystallites formed on a suitable support element by a several incipient wetness steps process and capable of withstanding at least 6 catalyst regenerations without significantly inhibiting it's catalytic activity in said reforming process, said Ni crystallites being [position] positioned in the inner surface of said suitable support element, said crystallites having an average maximum dimension measured in any one direction in the range of between [10] 5 and 1000 Å and a distribution on said support element of no more than 0.2 of said square meter ($m^2$) of exposed nickel/ square meter ($m\{2\}^2$) of support surface.

The paragraph beginning on page 6 line 16 has been revised as follows

Broadly the present invention also relates to a reforming process comprising reforming hydrocarbons in the presence of a catalyst in a reaction zone, said catalyst being Nickel (Ni) catalyst of discrete Ni crystallites formed on [said] a support element by a several step incipient wetness process, said crystallites having an average size measured in any one direction in the range of between [10] 5 and 1000 Å and a distribution on said support element of no more than 0.2 of said square meter of nickel exposed metal/ square meter of support selected from the group consisting of alumina and zeolite materials, recycling said catalyst to and from said reaction zone, regenerating between 10 and 100 % of the catalyst being recycled in a regeneration zone to provide a regenerated catalyst and returning said regenerated catalyst to said reaction zone

The paragraph beginning on page 9 line 24 has been revised as follows

It has been found that the size range characteristic dimension "l" for the Ni crystallites should be in the range of [10] 5 Å to 1000 Å and preferably are in the range of 150 to 250 Å These crystallites [104] 106 are distributed on the support [100] 102 so that there are no more than 0.2 $m^2$.of exposed nickel/ $m^2$ of support i.e. a minimum spacing between adjacent crystallites or maximum coverage of the surface support.

The paragraph beginning on page 10 line 1 has been revised as follows

The average range of size i.e. dimension l of the crystallites [104] 106 when α- alumina (the preferred form of support for steam reforming) is used as the support structure will generally be

in the range of 10Å to 1000 Å preferably 150Å to 250Å and for zeolite supports will normally be smaller than those for α alumina and generally will be in the range of 5Å to 100Å preferably 30Å to 70Å. It has been found that when the size and distribution of Ni crystallites are not within these ranges the resulting product has [catalyst] <u>catalytic</u> properties significantly inferior to those of the present invention.

**In the claims**

Claims 1 and 2 have been amended as follows

1. (Amended twice) A regenerable Nickel (Ni) catalyst [particularly suited] for a hydrocarbon reforming process, said catalyst comprising discrete Ni crystallites formed on a [suitable] support element by a several incipient wetness steps process <u>incorporating a plurality of Ni impregnation cycles, said catalyst being</u> [and] capable of withstanding at least 6 catalyst regenerations without significantly inhibiting it's catalytic activity in said reforming process, said Ni crystallites being positioned in the inner surface of said [suitable] support element, said crystallites having a crystallite characteristic dimension [measured in any one direction in the range] of between [10] <u>5</u> and 1000 Å and a [distribution] <u>dispersion</u> on said support element of no more than 0.2 square meter of exposed nickel/ square meter of support surface.

2. (Amended) A catalyst as defined in claim 1 wherein said support element is selected from the group [comprising] <u>consisting of</u> alumina and zeolite [and other suitable supports having equivalent physical characteristics].

Claims 4 and 5 have been amended as follows

4. (Amended twice) A catalyst as defined in claim 3 wherein said crystallite characteristic dimension [measured in any one direction] in the range of between 10 and 1000Å average size and a distribution on said support element of no more than $0.2m^2$ of exposed nickel $/m^2$ of said support surface.

5. (Amended twice) A catalyst as defined in claim 3 wherein said <u>characteristic dimension</u> [average size] is in the range of between 150 Å and 250 Å and a distribution on said support element of no more than 0.16 of said $m^2$ of exposed nickel / $m^2$ of said support surface.

Claims 9 to 23 inclusive have been amended as follows

9. (Amended twice) A catalyst as defined in claim 8 wherein said <u>characteristic dimension</u> [average size] is in the range of between 5 Å and 100 Å and a distribution on said support element of no more than 0.15 $m^2$ nickel exposed / $m^2$ of said support surface.

10. (Amended twice) A catalyst as defined in claim 8 wherein said <u>characteristic dimension</u> [average size] is in the range of between 10 Å and 70 Å of no more than 0.10 $m^2$ nickel exposed / $m^2$ of said support surface.

11 (Amended) A catalyst as defined in claim 9 wherein said zeolite is selected from the group consisting of NaY (sodium exchanged Y [type] zeolite) and USY (ultrastabilized Y [type] zeolite).

12. (Amended) A catalyst as defined in claim 10 wherein said zeolite is selected from the group ultrastabilized Y [type] zeolite.

13 (Amended) A catalyst as defined in claim 1 wherein said [suitable] support element has an average size in the range of between 5 and 200 microns.

14 Amended) A catalyst as defined in claim 1 wherein said [suitable] support element has an average size in the range of between 20 and 100 microns

15 (Amended) A catalyst as defined in claim 6 wherein said [suitable] support element has an average size in the range of between 5 and 200 microns.

16 (Amended) A catalyst as defined in claim 6 wherein said [suitable] support element has an average size in the range of between 20 and 100 microns.

17 (Amended) A catalyst as defined in claim 7 wherein said [suitable] support element has an average size is in the range of between 5 and 200 microns.

18 (Amended) A catalyst as defined in claim 7 wherein said [suitable] support element has an average size in the range of between 20 and 100 microns.

19 (Amended) A catalyst as defined in claim 11 wherein said [suitable] support element has an average size in the range of between 5 and 200 microns

20 (Amended) A catalyst as defined in claim 11 wherein said [suitable] support element has an average size in the range of between 20 and 100 microns.

21 (Amended) A catalyst as defined in claim 12 wherein said [suitable] support element has an average size in the range of between 5 and 200 microns.

22 (Amended) A catalyst as defined in claim 12 wherein said [suitable] support element has an average size in the range of between 20 and 100 microns.

23. (Amended twice) A reforming process comprising reforming hydrocarbons in the presence of a catalyst in a reaction zone, said catalyst being Nickel (Ni) catalyst of discrete Ni crystallites formed on [said] a support by a several step incipient wetness process incorporating a plurality of Ni impregnating steps, said crystallites having a maximum dimension [measures in any one direction] in the range of between [10] 5 and 1000 Å and a [distribution] dispersion on said support [element] of no more than 0.2 [of said] square meter of nickel exposed metal/ square meter of said support, said support being selected from the group consisting of alumina and zeolite materials, recycling said catalyst to and from said reaction zone, regenerating between 10 and 100 % of the catalyst being recycled in a regeneration zone to provide a regenerated catalyst and returning said regenerated catalyst to said reaction zone.

Please add new claims 26, 27 and 28 reading as follows

26. A catalyst as defined in claim 6 wherein said Ni crystallites are present in the amount of up to 20 wt%.

27. A catalyst as defined in claim 7 wherein said Ni crystallites are present in the amount of up to 20 wt%

28. A reforming process as defined in claim 25 wherein said Ni crystallites are present in the amount of up to 20 wt%

## Appendix

### Comparison of nickel alumina catalyst of van Looij et al and the catalyst of the present invention

*a) Nickel Loading*
*Nickel Loading= Weight of nickel (grams)/Weight of support (grams)*

- Nickel on Alumina-van Looij catalyst

Data from van Looij et al patent: 33.7 grams of $NiNO_3$ are used to impregnate 12 grams of alumina.

$$\text{WeightofNi} = 33.7\text{gramsNiNO}_3 \frac{58.69\text{gramsNi/mole}}{134.69\text{gramsNiNO}_3\text{/mole}} = 14.7\text{gramsNi}$$

Nickel Loading = 14.7grams Ni/ 12 grams alumina=1.22 grams Ni/grams alumina

**Theoretical Nickel Loading wt%=122 wt%**

Note that there is no data in van Looij et al patent about the efficiency of impregnation. However and given the method of impregnation used one should expect a high impregnation efficiency.

| Nickel Loading. Summary Table | Theoretical Nickel loaded | Efficiency of Impregnation |
|---|---|---|
| Van Looij et al, nickel on alumina | 122wt% | Estimated in the 80-90% range |
| Nickel on alumina catalyst of this invention | 1-20wt% with an optimum at 2.5% | 83% |

*b) Nickel Dispersion*

*Nickel Dispersion= Area of Nickel ($m^2$)/Area of support ($m^2$)*

To calculate nickel dispersion we have to introduce the following definitions:

- Area of one crystal= [Size of Crystal (m)]$^2$ x Exposed Crystal Faces
- Weight of 1crystal = [Size of Crystal (m)]$^3$ ($m^3$) x Nickel density (grams/$m^3$)
- Number of nickel crystals=Weight of Ni (grams)/weight on 1 crystal (grams)

- Area of Nickel ($m^2$)= Number of nickel crystals x area of 1 crystal ($m^2$)
- Area of support ($m^2$) = Specific Area ($m^2$/grams) x Weight of Support (grams)

- Using the above equations and the data of van Looij patent the following calculation can be developed

Data from patent: Ni crystals of 65A=6.5nm = $6.5\ 10^{-9}$ m

Area of 1 crystal= $[6.5\ 10^{-9}]^2 \times 5 = 2.11\ 10^{-16}\ m^2$

Weight of 1 crystal = $[6.5\ 10^{-9}]^3 \times 8190000 = 2.45\ 10^{-18}$ grams

Number of nickel crystals=14.68(grams)/2.45 $10^{-18}$(grams/crystal)= 6.0 $10^{18}$

Area of Nickel ($m^2$)= 6.0 $10^{18}$ x 2.11 $10^{-16}$ ($m^2$)= 1270 $m^2$

Area of support ($m^2$) = 100 $m^2$/g x 12 grams = 1200 $m^2$

**Nickel Dispersion= 1270 $m^2$ Ni/1200 $m^2$ support = 1.06**

| Nickel Dispersion-Summary Table | Dispersion $m^2$ Ni/$m^2$ support |
|---|---|
| Van Looij et al, Nickel on alumina | 1.06 |
| This invention-Nickel on alumina | 0.14 |